Competition between the BCS superconductivity and ferromagnetic spin fluctuations in $MgCNi_3$

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The low temperature specific heat of the superconductor $MgCNi_3$ and a non-superconductor $MgC_{0.85}Ni_3$ is investigated in detail. An additional contribution is observed from the data of $MgCNi_3$ but absent in $MgC_{0.85}Ni_3$, which is demonstrated to be insensitive to the applied magnetic field even up to 12 Tesla. A detailed discussion on its origin is then presented. By subtracting this additional contribution, the zero field specific heat of $MgCNi_3$ can be well described by the BCS theory with the gap ratio (Δ/k_BT_c) determined by the previous tunneling measurements. The conventional s-wave pairing state is further proved by the magnetic field dependence of the specific heat at low temperatures and the behavior of the upper critical field.

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Since the discovery of the new intermetalic perovskite superconductor $MgCNi_3$ [1], plenty of efforts have been focused on the superconducting pairing symmetry in this material because its conduction electrons are derived predominantly from Ni which is itself a ferromagnet [2, 3, 4, 5]. However, up to now, there is still not a consensus on this issue. The measured penetration depth [6], critical current behavior [7] and earlier tunneling spectra [8] suggested an unconventional superconductivity, the later tunneling data [9] supported the s-wave pairing symmetry and gave a reasonable interpretation on the contradiction to the result in Ref[8]. The s-wave pairing has also been demonstrated by the ^{13}C NMR experiments [10] and the specific heat measurements [1, 8, 11, 12, 13, 14]. To our knowledge, all the previous reports on the specific heat of $MgCNi_3$ [1, 8, 11, 12, 13, 14] were characterized in the framework of a conventional phonon-mediated pairing. However, there is an obvious deviation of the experimental data from the prediction of BCS theory in the low temperature [8, 15], i.e., the entropy conservation rule is not satisfied. Such deviation has been interpreted by the presence of unreacted Ni impurities in Refs[8, 15], whereas it is still prominent in the samples without Ni impurities [14]. On the other hand, strong spin fluctuations have been observed in $MgCNi_3$ by NMR experiment [10], which is suggested to be able to severely affect the superconductivity in $MgCNi_3$ [2, 5, 10, 11, 16] or even induce some exotic paring mechanism [2]. Consequently, the behavior of the specific heat will inevitably be changed by the spin fluctuations. Therefore, before a real pairing mechanism being concluded from the specific heat data, we have to carefully investigate how the ferromagnetic spin fluctuations contribute to the specific heat of $MgCNi_3$.

In this work, we elaborate on the specific heat (C) of MgC_xNi_3 system both in normal state and superconducting state. A low temperature upturn is clearly distinguished in the C/T vs T^2 curves and found to be insensitive to the applied magnetic field. By doing some quantitative analysis, we present the evidence of most possible mechanisms responsible for this upturn. After subtracting this additional contribution, a well defined BCS-type electronic specific heat is extracted. The temperature dependence of the upper critical field and the field dependence of the low temperature specific heat also supports such conventional BCS superconductivity in $MgCNi_3$. These analyses indicate that although the spin fluctuations may suppress the pairing strength in $MgCNi_3$, the superconductivity is certainly not induced by any exotic mechanism.

Poly-crystalline samples of MgC_xNi_3 were prepared by powder metallurgy method. Details of the preparation were published previously [17]. The superconductor $MgCNi_3$ has a T_c of $6.7K$ and the non-superconductor $MgC_{0.85}Ni_3$ was synthesized by continually reducing the carbon component until the diamagnetism was completely suppressed. The heat capacity data presented here were taken with the relaxation method [18] based on an Oxford cryogenic system Maglab in which the magnetic field can be achieved up to 12 Tesla. Details of the sample information and the measurements can be found in recent report [11]. It should be emphasized here that the Cernox thermometer used for calorimetry has been calibrated at 0, 1, 2, 4, 8 and 12 Tesla, and the calibration for the intermediate fields is performed by an interpolation using the result of the adjacent fields. Therefore, any prominent field dependence of the specific data should reflect the intrinsic properties of the measured sample.

In general, the low temperature specific heat $C(T,H)$ of a superconductor consists four main contributions by neglecting the component of the nuclear moments [20, 29], each has a different dependence on T and two of which depend on H, also in different ways,

$$C(H,T) = C_{mag}(H,T) + C_{DOS}(H,T) + \gamma_0 T + C_{ph}(T) \quad (1)$$

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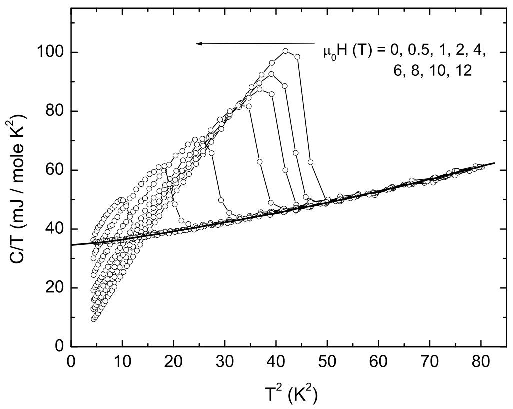

FIG. 1: The low-temperature specific heat of $MgCNi_3$ at various magnetic fields from 0T to 12T. The thick solid line denotes the universal background including all the normal state data for different fields.

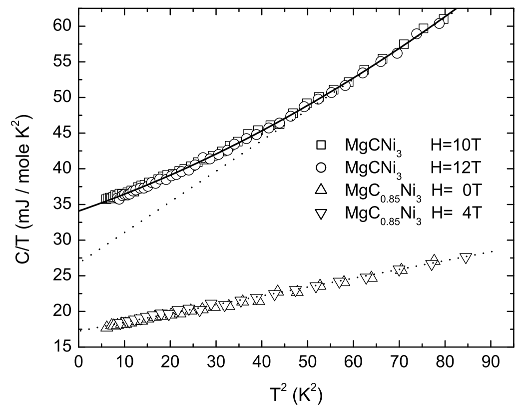

FIG. 2: A plot of C/T vs T^2 for $MgCNi_3$ and $MgC_{0.85}Ni_3$ in different magnetic fields. The data of $MgC_{0.85}Ni_3$ can be well defined by a straight line while that of $MgCNi_3$ remarkably deviate the linearity. The dashed line is a linear extrapolation of the high temperature data of $MgCNi_3$ and the upper solid line is the theoretical fit considering higher order phonon contributions.

where $\gamma_0 T$ represents a spare zero-field linear term and C_{ph} is due to the lattice or phonon contribution. The Debye phonon specific heat $C_{ph} = \beta T^3$ can usually describe the lattice contribution at low temperatures. However, the departure from T^3-law has often been observed, which is due to the fact that the density of modes of the phonon in real solid does not follow the assumed ω^2-law, here ω is the angular frequency of a harmonic wave associated with the lattice vibration. In such case, the deviations may be expanded in higher order terms such as T^5, T^7, etc.. The H-dependent terms in Eq. (1), i.e., $C_{mag}(H)$ and $C_{DOS}(H)$, are the contributions associated with magnetism and the electronic density of states (DOS), respectively. If there is no magnetism associated contribution, the normal state specific heat at low temperature can be approximatively described as $C_n(T) = \gamma_n T + \beta T^3$ in the framework of metal theory. Therefore, a linear relation can be obtained by plotting the normal state data as $C_n(T)/T$ vs T^2, and its intercept and slope correspond to γ_n and β, respectively.

The low temperature specific heat at various magnetic fields up to 12 Tesla is plotted as $C(T)/T$ vs T^2 in Fig. 1. Two important features should be emphasized here. First, all the normal state data at various magnetic fields merge into one [11], which is consistent with the results reported by other groups [14, 15]. Second, this common normal state background remarkably deviates from the linear relation as discussed above. In Fig. 2, only the normal state data are re-plotted in an magnified scale. In order to survey the normal behavior at very low temperature, the magnetic field up to 12 Tesla was applied in measurements, which exceeds the highest upper critical field of our sample and is 4 Tesla higher than that used by other groups [14, 15]. The specific heat of the non-superconductor $MgC_{0.85}Ni_3$ is also presented in Fig. 2 as a comparison. It is obvious that the perfect linear relation of $C(T)/T$ vs T^2 is satisfied for $MgC_{0.85}Ni_3$, which is a striking contrast with the case of $MgCNi_3$.

The obvious upturn in the low temperature $C(T)/T$ vs T^2 curves of $MgCNi_3$ can not be associated with Ni impurites since the X-ray diffraction pattern shows no indication for Ni impurites [11]. To say the least, if there is still extreme small content of Ni impurites leading to the prominent low temperature upturn of C/T, the field dependence of its specific heat should also be obvious, which is clearly inconsistent with our experimental results. Moreover, if this upturn is due to the excess free Ni in $MgCNi_3$, it should also be observed in $MgC_{0.85}Ni_3$ because of the similar process of synthesizing these two samples. Quantitatively, taking the data from references [21, 22] yields for 10% of superfluous Ni an upturn which is at least two orders of magnitude smaller than the observed one. Therefore, the contribution of the excessive Ni can be neglected comparing with the whole specific heat. Furthermore, the possible Schottky anomaly is presented in Fig.3, its field dependence is obviously too strong to compare with our experimental result (nearly field independent).

It is found that this upturn can be well fitted if the above mentioned T^5 term is considered (see the upper solid line in Fig. 2). In other words, the departure from the T^3 behavior may be due to the non-Debye phonon DOS, which is consistent with the notable difference of the Debye temperature between $MgCNi_3$ and $MgC_{0.85}Ni_3$ [11]. If the electron-phonon coupling is indeed the origin of superconductivity in $MgCNi_3$, it is reasonable to associate the disappearance of superconductivity in $MgC_{0.85}Ni_3$ with the remarkable difference of its phonon DOS from that of $MgCNi_3$. However, some careful work is needed to understand such obvious difference of phonon structure between these two samples, since they have similar crystal lattices and chemical components.

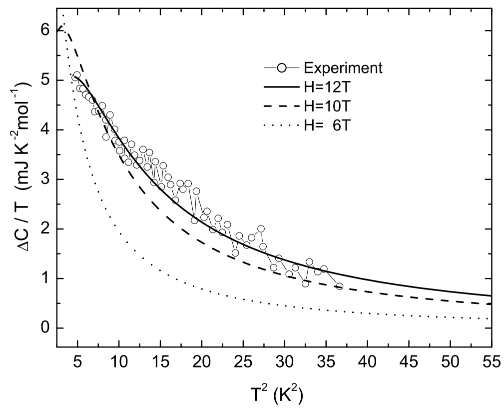

FIG. 3: Comparison between the field-insensitive low-temperature upturn in the specific heat of $MgCNi_3$ ($\Delta C = C - \gamma T - \beta T^3$) and the calculated field dependent Schottky anomaly.

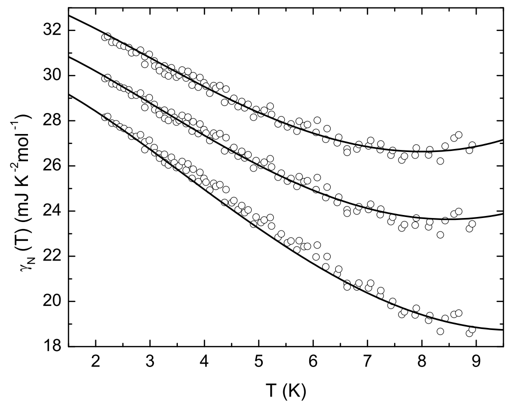

FIG. 4: Electronic specific heat $\gamma_n(T)$ versus T of $MgCNi_3$ in the normal state (three different β-values are selected in order to avoid artificial errors.). The solid lines are theoretical fits to spin-fluctuation model. All the curves except the top two are shifted downwards for clarity.

Another possible explanation of the above mentioned low temperature upturn is the existence of strong spin fluctuations due to the higher DOS at fermi energy ($N(E_F)$) of $MgCNi_3$ than that of $MgC_{0.85}Ni_3$ [11], consequently, the coupling between the electrons and spin fluctuations in $MgCNi_3$ should also be stronger. The ferromagnetic spin fluctuations have been demonstrated by NMR experiments [10]. Doniach and Engelshberg [23] and Berk and Schrieffer [24] showed that the absorption and re-emission of spin fluctruations renormalizes the electronic self-energy, leading to an enhanced effective mass at low temperatures. This effect manifests itself as a low-temperature enhancement of the electronic specific-heat coefficient, λ_{sf}, which depends on temperature as $T^2 ln(T/T_{sf})$ (here T_{sf} is the characteristic spin-fluctuation temperature) at low temperature. Considering the presence of ferromagnetic spin fluctuations, the normal state specific heat of MgC_xNi_3 can be expressed as follows,

$$C_n(H=0,T) = A[1+\lambda_{ph}+\lambda_{sf}(T)]T + \gamma_0 T + \beta T^3 \quad (2)$$

where βT^3 are the contributions of phonon excitations, $\lambda_{sf}T$ and $\lambda_{ph}T$ represent the contributions of effective mass renormalization due to the electron-spin fluctuation coupling and the electron-phonon coupling, respectively, and A is a constant correlated with $N(E_F)$. It can be seen from Eq. (2) that the deviation from the linear dependence of $C(T)/T$ on T^2 is due to the temperature dependence of λ_{sf}. Moreover, Béal-Monod, Ma, and Fredkin [25] have estimated the shift $\delta C/T$ caused by an applied field H to be

$$\delta C/T \approx 0.1(\frac{\mu H}{k_B T_{sf}})^2 \frac{S}{lnS} \quad (3)$$

where S is Stoner factor. Eqs. (2) and (3) indicate that the possible magnetic field dependence of the normal state specific heat is completely determined by the spin fluctuations. For simplicity, Eq. (2) can be rewritten as $C_n(H=0,T) = \gamma_n(T)T + \beta T^3$, in which $\gamma_n(T) = A[1+\lambda_{ph}+\lambda_{sf}(T)+\gamma_0/A]$. Therefore, the $\gamma_n \sim T$ relation directly reflects the temperature dependence of λ_{sf}. In Fig. 4, we present the determined $\gamma_n(T)$ by selecting various β-values. Fitting the $\gamma_n(T)$ relations to the formula of $A(1+BT^2 ln(T/T_{sf}))$ yields T_{sf} varying from 13 to 16K. By inserting the determined T_{sf}, calculated Stoner factor S [2] and the highest field value in our measurements into Eq. (3), we can estimate the shift $\delta C/T$ caused by the applied field to be less than 2%, which is in agreement with our experimental results. However, if this explanation is correct, we must understand the collapse of the entropy conservation around T_c caused by considering such additional electronic specific heat, as discussed below. Therefore, the specific-heat contribution of the spin fluctuations themselves may be another candidate responsible for the low temperature upturn in specific heat of $MgCNi_3$.

Despite the true mechanism of the low-temperature upturn of C/T, this additional specific heat contribution should be regarded as a part of the normal-state background of the superconducting specific heat below the upper critical field $H_{c2}(T)$. In earlier analysis to the specific heat data [8, 11, 12], this additional part of background has been neglected more or less below $H_{c2}(T)$. We point out here that neglecting this additional contribution will lead to the collapse of the entropy conservation as reported in references [8, 15]. This opinion is motivated by the subsequent analysis. As shown in Fig. 5(a), the normal state background (as shown in Fig. 2) has been subtracted from the zero-field specific heat data, the entropy difference $\Delta S(T) = \int_0^T dT(\Delta C/T)$ is presented in the inset of Fig. 5(a), here $\Delta C = C_{H=0} - C_n$. It is found that the entropy conservation is then well satisfied, indicating that the remainder is the contribution of superconducting state. Such analysis has also been ap-

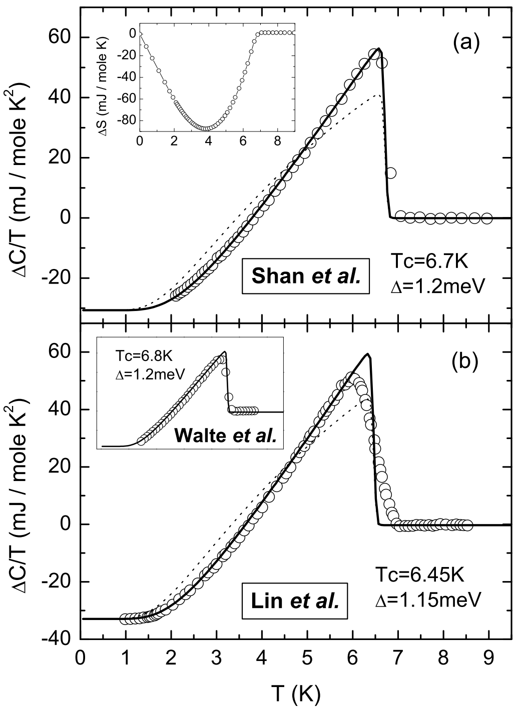

FIG. 5: Fitting the specific heat data ($C_{H=0} - C_n$) measured by different groups [11, 14, 15] to BCS model. The experimental data are denoted by open circles and the fits to α-model are denoted by solid lines. The dotted lines are fits to the original BCS model. The inset in (a) is the entropy difference by integration of $\Delta C/T$ as presented in (a).

plied on the data measured by Wälte *et al.* [14] and Lin *et al.* [15], respectively, the entropy is also conserved and the low temperature anomaly as mentioned in Ref.[15] completely vanishes.

When the superconductivity in $MgCNi_3$ is investigated, the spin fluctuations can not be neglected because it may compete with superconductivity [5, 11] or even lead to an exotic pairing mechanism other than the conventional s-wave [2]. If the phonon-mediated pairing wins in competing with the spin fluctuations and hence the effect of the spin fluctuations only suppress the electron-phonon coupling (or pairing-strength) [11], the so-called α-model [26] based on BCS theory should be a good choice to describe the measured thermodynamic parameters. Comparing with the original BCS-model, the only adjustable parameter in this α-model is the gap ratio $\Delta(0)/k_BT_c$. This model has been successfully applied to strong coupling systems such as Pb and Hg. Then we try to fit the superconducting part of the zero-field specific heat to the α-model, the results are presented in Fig. 5. All the data can be well described by this revised BCS model with the best fitting parameters (i.e., Δ and T_c) listed in Table. I. These fits yield a gap ratio $\Delta(0)/k_BT_c \approx 2.06$, corresponding to the maximum gap $\Delta(0) \approx 1.2$meV which is in good agreement with our previous tuneling measurements [9]. From the

TABLE I: Fits to BCS model for zero-field specific heat.

Groups	Δ (meV)	T_c (K)	$2\Delta/k_BT_c$
Shan *et al.* [11]	1.20	6.70	4.15
Wälte *et al.* [14]	1.20	6.80	4.10
Lin *et al.* [15]	1.15	6.45	4.14

above discussions, we can conclude that the coexistence and competition of spin fluctuations and phonons does not change the phonon-mediated pairing mechanism of $MgCNi_3$.

In order to further verify this picture, we investigate the field dependence of the low temperature specific heat of $MgCNi_3$. It is known that the electronic specific heat in magnetic fields can be expressed by $C_{el}(T,H) = C_{el}(T,H=0) + \gamma(H)T$. The magnetic field dependence of $\gamma(H)$ is associated with the form of the gap function of the superconductor. For example, in a superconductor with line nodes in the gap function, the quasiparticle DOS ($N(E)$) rises linearly with energy at the Fermi level in zero field, $N(E) \propto |E - E_F|$, which results in a contribution to the specific heat $C_{DOS} = \alpha T^2$ [27]. In the mixed state with the field higher than a certain value, the DOS near the Fermi surface becomes finite, therefore the quadratic term $C_{DOS} = \alpha T^2$ will disappear and be substituted by the excitations from both inside the vortex core and the de-localized excitations outside the core. For d-wave superconductors with line nodes in the gap function, Volovik *et al.* [28] pointed out that in the mixed state, supercurrents around a vortex core cause a Doppler shift of the quasi-particle excitation spectrum. This shift has important effects upon the low energy excitation around the nodes, where its value is comparable to the width of the superconducting gap. For $H >> H_{c1}$, it is predicted that $N(E_F) \propto H^{1/2}$ and $C_{DOS} = \Delta\gamma(H)T = ATH^{1/2}$ at low temperatures [28]. This prediction has been well proved for hole doped cuprates [29]. Whereas in a conventional *s*-wave superconductor, the specific heat in the vortex state is dominated by the contribution from the localized quasi-particles in the vortex cores. From the Bogoliubov equations assuming noninteracting vortices, the DOS associated with the bound excitations is derived as $N(E) \propto B(H)$ [30] hence the contribution of the vortex cores to the specific heat is $C_{DOS} \propto B(H)T$ [31, 32]. It is also theoretically derived that the experimentally observed downward curving $C(H)$ is caused by the flux line interactions near H_{c1} and the possible expansion of the vortex cores [31, 32]. According to the above discussions, the specific heat coefficient $\gamma(H)$ of conventional *s*-wave superconductor should linearly depend on the magnetic field well above H_{c1}.

Fig. 6 shows the field dependence of $\gamma(H) - \gamma(0)$ of $MgCNi_3$ below 3K. The data reported by different groups [11, 14, 15] merge into each other by timing a prefactor A close to unity. It is found that γ linearly depends on

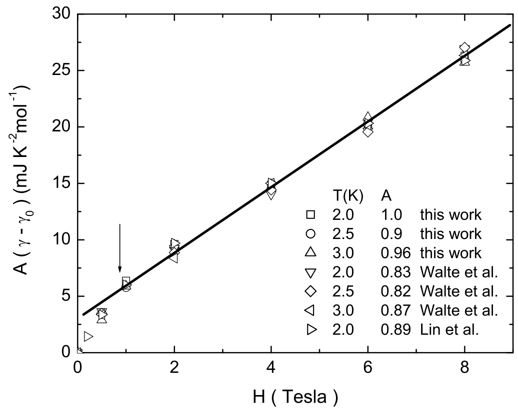

FIG. 6: The magnetic field dependence of the specific heat coefficient $\gamma(H) - \gamma(0)$ at low temperatures.

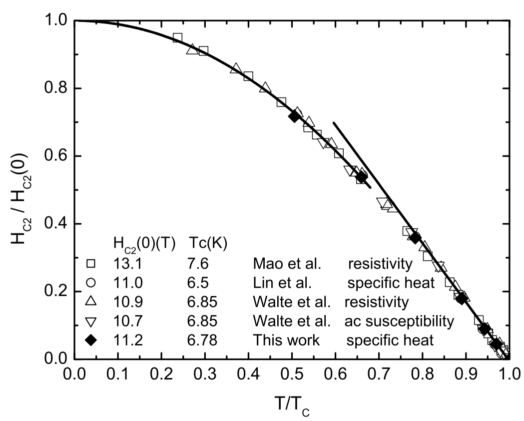

FIG. 7: The comparison of the temperature dependence of the upper critical field $H_{c2}(T)$ with the BCS-like descriptions in lower and higher temperature limits.

H above 0.5T and persists up to 8T which is close to the upper critical field of $MgCNi_3$. The legible linearity of $\Delta\gamma \sim H$ relation at higher field and its negative curvature below 0.5T are in good agreement with the above mentioned behaviors of conventional s-wave superconductors. It may be argued that the low temperature limit of about 2K in our measurements is not low enough to distinguish the d-wave's $\Delta\gamma \sim H^{1/2}$-law. However, it should be emphasized that the observed $\Delta\gamma \sim H$ relation is nearly universal at low temperatures below upper critical field, which is very similar to the behavior of V_3Si [32], a typical conventional s-wave superconductor.

Finally, we compare the temperature dependence of the upper critical field $H_{c2}(T)$ with the prediction of BCS theory in which the $H_{c2}(T)$ can be expressed as follows,

$$H_{c2}(T) \approx 1.74 H_c(0)(1 - T/T_c) \quad (T_c - T \ll T_c) \quad (4a)$$

$$H_{c2}(T) \approx H_{c2}(0)[1 - 1.06(T/T_c)^2] \quad (At\ low\ T) \quad (4b)$$

As shown in Fig. 7, the best fitting to BCS model is denoted by solid lines. At lower temperature, the experimental data can be well described by Eq.(4b). For the higher temperature near T_c, a prefactor of 1.65 is obtained instead of the theoretical prediction of 1.74 as expressed in Eq.(4a). Nonetheless, the BCS model is still a preferred description for $H_{c2}(T)$ of $MgCNi_3$ considering the stronger electron-phonon coupling and the presence of ferromagnetic spin fluctuations.

In summary, we have investigated the specific heat data of MgC_xNi_3 system. A remarkable field independent contribution is found in $MgCNi_3$, reflecting the departure of normal-state specific heat from T^3-law. By removing this contribution, the zero-field data is well described by the α-model (a slightly revised BCS model). The conventional s-wave superconductivity is further supported by the linear field dependence of specific heat coefficient $\gamma(H)$ and the BCS-like temperature dependence of upper critical field $H_{c2}(T)$. It is then concluded that, although electron-magnon (spin fluctuations) coupling coexists and competes with electron-phonon coupling effect in $MgCNi_3$, it only acts as pair breakers while does not induce a new exotic superconductivity.

Note added: Most recently, the carbon isotope effect in superconducting $MgCNi_3$ observed by T. Klimczuk and R.J. Cava indicates that carbon-based phonons play a critical role in the presence of superconductivity in this compound [33].

Acknowledgments

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